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Naturally Occurring Radium-226 and Radium-228 in Water Supplies of Michigan

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Radium (Ra) is a naturally occurring radioactive metal which exists in several isotopic forms. These isotopes are formed by decay of uranium and thorium in nature. The predominant radium isotopes in groundwater are Ra^{226} and Ra^{228} (ATSDR, 1990). Radium is a known (Croup A) carcinogen. This means there is sufficient evidence from epidemiological studies to support a causal association between exposure to radium and cancer in humans (U.S. EPA, 1986a; U.S. EPA, 1996a). Absorbed radium is stored in bone tissues. The half-life of Ra^{226} and Ra^{228} is 1600 and 5.75 years, respectively. Chronic human exposure to Ra^{226} is associated with occurrence of bone sarcomas and carcinomas of the perinasal sinuses and mastoid air cells (commonly called head carcinomas). Chronic exposure to Ra^{228} is only associated with bone sarcoma (ATSDR, 1990; U.S. EPA, 1991). The relative effectiveness of Ra^{226} and Ra^{228} to cause bone sarcoma in humans is estimated to be 1:2.5 (Rowland et al., 1978). Exposure to high levels of radium causes several adverse health effects such as benign bone growth, osteoporosis, growth retardation, kidney disease, liver disease, tissue necrosis, cataract, anemia, immunological depression, and death (ATSDR, 1990; U.S. EPA, 1991).

The National Interim Primary Drinking Water Regulation for combined Ra^{226} and Ra^{228} was set at 5 pCi/L (U.S. EPA, 1976; U.S. EPA, 1996b). The proposed Drinking Water Maximum Contaminant Levels for Ra^{226} and Ra^{228} were set at 20 pCi/L by U.S. EPA (1991). However, these proposed standards were later withdrawn by the agency in 1993 for further review (U.S. EPA, 1997).

Chemical analyses of water supply wells in Saginaw County had previously indicated the presence of low levels of radium (8-9 pCi/L) in groundwater (Michigan Department of Environmental Quality files, 1982). The objective of this paper is to report the data collected for Ra^{226}, Ra^{228}, and total dissolved solids in groundwater samples obtained from four counties (Bay, Midland, Saginaw, and Tuscola) of the Saginaw Valley area in Michigan. Also discussed are the potential health risks related to Ra^{226} and Ra^{228}.

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MATERIALS AND METHODS

The aquifers in this study area could be divided into two distinct geological units. These are consolidated bedrock of Mississippian-Pennsylvanian age, overlain by unconsolidated sediments of Pleistocene age. The consolidated bedrock unit consists of interlayed sandstone and shale. Drinking water wells completed into this unit have an open borehole below the surface casing and are referred to as rock wells in this paper. The unconsolidated sediments consist predominantly of clay with minor sand and gravel layers. Drinking water wells completed into this unit have a screen attached to the bottom of the surface casing and are referred to as screened wells in this paper. Water samples from 149 wells were collected from Bay, Saginaw, parts of Midland, and Tuscola counties of Michigan. Wells were selected so that each township had two to four wells sampled. Additionally, it was attempted to sample one rock and one screened well per township. Selection of homes to be sampled was based upon the presence of a verifiable well construction record showing the method of drilling, geologic formation penetrated, casing diameter, and depth. The above information was verified with the homeowner at the time of sampling a well.

The U.S. Environmental Protection Agency's (EPA) Eastern Environmental Radiation Laboratory Facility in Montgomery, Alabama, analyzed water samples for Ra^{226} and Ra^{228} using the Krieger and Whittaker (1980) procedures for measurement of radioactivity in drinking water. The water samples were shipped in one gallon plastic containers.

For determination of total dissolved solids, water samples were collected in 500 ml plastic bottles and were shipped to the State Drinking Water Laboratory, Lansing, Michigan. The total dissolved solids were determined by using the U.S. EPA approved procedure number 160.1. The statistical analyses of the data were performed as outlined by Steel et al., (1997). The radium data as expected for naturally occurring elements (Rose et al., 1979) was highly skewed. Therefore, all radium data points were transformed to normalcy by logarithm to base 10 prior to statistical analyses. Comparisons for radium data between counties and between well types were made by using Student's-t-test. The risk assessment formulations for Ra^{226} and Ra^{228} were adopted from U.S. EPA (1986b).

RESULTS AND DISCUSSION

The concentrations of Ra^{226} and Ra^{228} in some groundwater wells in Michigan are shown in Tables 1 and 2. The highest concentrations of Ra^{226} and Ra^{228} were observed in Bay County, followed by Midland, Saginaw, and Tuscola counties, respectively (Table 1). About 37% of the samples analyzed exceeded 5 pCi/L level. The concentrations of Ra^{226} and Ra^{228} were higher ($P<0.01$) in samples obtained from rock wells as compared to the screened wells (Table 2). The

Table 1. Concentration of Ra^{226} and Ra^{228} in groundwater wells in Saginaw Valley area of Michigan' (Mean ± SE[2]).

County	Well Type	*n*	Ra^{226}	Ra^{228}	$Ra^{226+228}$	$Ra^{226+228}$ All wells[3]
			----------pCi/L----------			
Bay	R[4]	28	7.2±2.0	5.6± 2.0	12.8±4.0	9.7 ± 2.6[a]
	S[5]	16	2.4±1.0	1.8±0.4	4.2±1.3	
Midland	R	17	4.0±0.8	4.6+1.0	8.6+1.7	6.3 ± 1.4[a]
	S	7	0.2+0.0	0.6±0.2	0.8±0.2	
Saginaw	R	47	2.3±0.4	2.6±0.5	4.9±0.9	4.6 ± 0.8[b]
	S	14	1.2+0.4	2.1±1.0	3.3±1.4	
Tuscola	R	16	1.7±0.4	2.3±0.8	4.0±1.2	3.4 ± 1.0[b]
	S	4	0.4±0.3	0.5±0.4	0.9±0.7	

[1]Arranged by county and well type
[2]Standard error
[3]All wells include both rock and screen wells tested in a county
[4]Rock wells
[5]Screened wells
[a,b]Values bearing different subscripts are statistically different (P<0.05)

Table 2. Concentration of Ra^{226} and Ra^{228} in groundwater wells in Saginaw Valley area of Michigan' (Mean ± SE[2]).

Wells	*n*	Ra226	Ra^{228}	$Ra^{226+228}$
		----------pCi/L----------		
Rock wells	108	3.7±0.6	3.7±0.6	7.4±1.2[a]
Screened wells	41	1.4±0.4	1.6±0.4	3.0±0.7[b]
All wells	149	3.1±0.5	3.1±0.5	6.2±0.9

[1]Arranged by well type
[2]Standard error
[a,b]Values bearing different subscripts are statistically different (P<0.01)

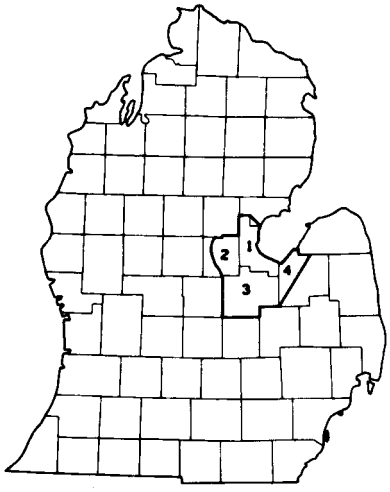

Figure 1. Localization of naturally occurring Ra^{226} and RA^{228} in groundwater in Michigan counties: 1) Bay, 2) Midland, 3) Saginaw and 4) Tuscola.

arithmetic mean concentrations of Ra^{226} and Ra^{228} in all the wells (n=149) were 3.1 ± 0.5 for both the radioisotopes. The reported nationwide arithmetic means (excluding California) are 1.7 and 3.6 pCi/L for Ra^{226} and Ra^{228}, respectively (Aieta et al., 1987).

The results of the levels of Ra^{226} and Ra^{228} in groundwater were considered too low to cause any acute adverse effect in humans, however, chronic exposure to low levels of Ra^{226} and Ra^{228} that naturally exists in this study area may lead to development of bone sarcoma and head carcinoma (ATSDR, 1990, U.S. EPA, 1991).

The naturally occurring radium in groundwater represented about 1,800 square miles of area (Figure 1). The total human population in this area was 405,872 persons (Michigan Department of Management and Budget, 1997). Statewide about 43% of Michigan's human population uses water from private wells for drinking (United States Geological Survey, 1992). Due to prevalence of municipal water supply lines (Lake Huron source) in the study area, about 22% of the residents in this study obtain their drinking water from private wells (Michigan Department of Environmental Quality Files, 1992). Therefore, it was calculated that 89,292 persons (405,872 x 0.22) are exposed to naturally occurring radium in Michigan. As only 37% of the samples exceeded SpCi/L level, it was computed that a total of 33,038 persons (89,292 x 0.37) are exposed to combined Ra^{226} and Ra^{228} at a concentration of ≥ 5pCi/L.

The risk assessment formulations adopted from U.S. EPA (1986b) are shown in

Table 3. Estimated lifetime risk levels.

RA^{226} pCi/l	Lifetime Risk Level*	Ra^{228} pCi/L	Lifetime Risk Level*
10	1×10^{-4}	20	1×10^{-4}
5	5×10^{-5}	10	5×10^{-5}
1	1×10^{-5}	2	1×10^{-5}
0.1	1×10^{-6}	0.2	1×10^{-6}

*Adopted from U.S. EPA (1986b).

Table 3. It appears that as a result of lifetime ingestion of 2L of drinking water The per day contaminated with Ra^{226} and Ra^{228} at 5 pCi/L, the number of excess cancer cases may vary from zero to five in a human population of 100,000 (Table 3). The development of excess cases of bone sarcoma or head carcinoma in a population of 33,038 persons exposed to combined Ra^{226} and Ra^{228} at 5 pCi/L may vary from zero to two. Since risk assessment models used to drive drinking water standards are conservative, health risk probability estimates mentioned here are also likely to be conservative. The acceptable risk level for environmental carcinogens in Michigan (State of Michigan, 1995) is one case of excess cancer per 100,000 persons (1×10^{-5}).

The background rate of cancer in America from all sources, including smoking, is approximately one case per four persons (American Cancer Society, 1992; Sidhu and Sidhu, 1996; MacDonell et al., 1998). Therefore, in a population of 100,000 people, 25,000 would develop some kind of cancer in their lifetime. If the same 100,000 persons daily drank water contaminated with the combined Ra^{226} and Ra^{228} at 5 pCi/L over their lifetime, it is estimated that no more than five additional persons may develop cancer. In other words 25,005 cases of cancer may occur in that population.

The mean concentrations of total dissolved solids (TDS) were 2349 ± 187,2022 ± 223, and 2259 ± 148 mg/L in water samples collected from rock wells, screened wells, and all the wells, respectively. The concentration of total dissolved solids was positively correlated ($p<0.001$) with the combined concentration of Ra^{226} and Ra^{228} in all wells (Figure 2). The coefficient of correlation (r) was 0.87. This finding is consistent with results reported in the literature for other high TDS waters with dissolved radium. Bloch and Key (198 1) and Kraemer and Reid (1984) both noted the direct correlation between TDS and radium concentration in oil-field brines. While oil and gas development is present in the study area, this is not believed to be the source of the anomalous radium due primarily to the wide spread geologic and geographic distribution of high TDS waters in the study area (Long et al., 1986; Rheaume, 1991) and historical records of Michigan's water quality (Lane, 1899;

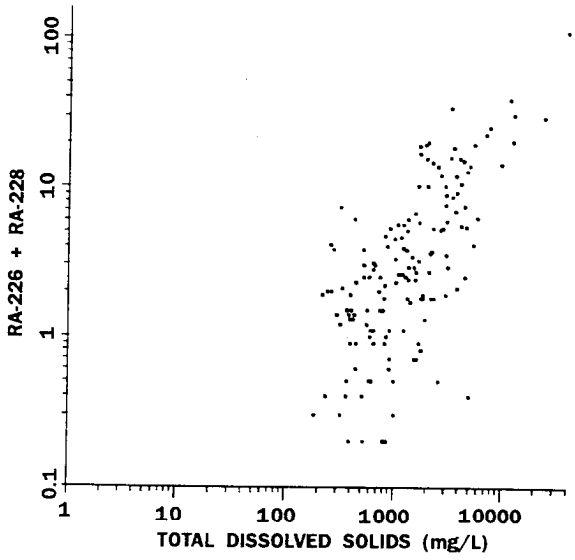

Figure 2. Relationship between naturally occurring $Ra^{226+228}$ and total dissolved solids in groundwater in Michigan.

Cummings, 1980). This same argument can be made for other anthropogenetic sources such as road salting during the winter months.

The water sample results were communicated to each residential well owner. We fully explained radium related potential health risks, estimated cancer risks, and regulation for combined total Ra^{226} and Ra^{228}.. The potential health risks were also explained to the public via telephone calls and press interviews.

The methods available for removing radionuclides from drinking water have been reviewed by Reid et al., 1985. The processes that are effective in removing radium from drinking water include lime softening, cation exchange, reverse osmosis, and selective absorption. The waste from those processes should be handled and disposed of according to radioactive waste disposal guidelines of the Michigan Department of Environmental Quality. All this information about the treatment of contaminated water and disposal of radioactive waste was provided to the public.

In conclusion, the occurrence of Ra^{226} and Ra^{228} in groundwater is spread over 1,800 square miles of area in Michigan. The arithmetic mean concentrations of Ra^{226} and Ra^{228} for all the wells (n=149) were 3.1 ± 0.5 pCi/L for both of the isotopes. Thirty-seven percent of the water samples exceeded the combined Ra^{226} and Ra^{228} standard set at 5 pCi/L. About 33,038 persons are likely to be exposed to the combined Ra^{226} and Ra^{228} at a concentration of ≥ 5 pCi/L. The concentrations of Ra^{226} and Ra^{228} were considered too low to cause any acute

adverse effect in humans. Chronic exposure to low levels of Ra^{226} and Ra^{228} may cause bone sarcoma and head carcinoma in humans. The estimated lifetime risk is zero to five cases of excess cancers in a population of 100,000 people. The estimated risk is zero to two cases of excess cancers for the total population (405,872 persons) of the study area. This information was shared with the affected residents in the study area.

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